Atomic Radiative Lifetimes Measured by Pulsed Laser Spectroscopy in the UV/VUV Spectral Region

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Abstract

Experimental lifetime measurements on free atoms and ions performed by using pulsed laser excitation in the ultraviolet and vacuum-ultraviolet spectral regions are presented. Different methods of laser pulse generation, atomization and light detection are described. A short summary of results obtained is also given. Future possibilities in determination of lifetimes and transition probabilities in the short wavelengths region are discussed.

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1. Introduction

The structure of atoms and their interaction with the surrounding are reflected in their emission or absorption spectra. Intensities of spectral lines give information not only on atomic properties but also on their concentration in the observed object. The line strengths, oscillator strengths and transition probabilities are radiative constants of atoms related to transitions between different energy states and they govern the intensities of spectral lines. These quantities can be interrelated through simple expressions by using atomic constants describing multiplicities and energy separations. The radiative constants are discussed in many textbooks on atomic physics, for example [1-3], and reviews on the experimental methods of their determination have been published; see e.g. [4-7].

Transition probabilities in fact provide a particularly severe test of atomic calculations, because they are rather sensitive to the wavefunctions of both levels involved in the transition and to the approximations used, especially when electron correlations and relativistic effects are considered. On a more fundamental level, measurements of transition probabilities are also being used to explore the non-conservation of parity predicted by the unified theories of the weak and electromagnetic interaction; here, highly forbidden transitions must be tested for an admixture of allowed transition probabilities [8,9].

Besides the basic interest in atomic quantities, the values of transition probabilities are used in different fields of research and technology: astrophysics, atmospheric physics, plasma physics, thermonuclear-fusion research and development of laser systems. The evaluation of atomic concentrations can frequently be performed from observed light intensities, when direct concentration measurements are not possible. A large data compilation has been performed at NIST [10-13]. Presently, a great part of the data collection and systematization is performed for astrophysical needs within the international Opacity Project and the Iron Project [14,15]. Since 1990, when the Goddard High-Resolution Spectrograph on the Hubble Space Telescope started to work, the demand for transition probabilities for VUV spectral lines dramatically increased [16,17].

Transition probabilities have mainly been determined from calculations and to a much smaller extend from experiments [18]. Accurate experimental data are needed for checking of theoretical models and methods. Furthermore, in many cases, especially for complex heavy atoms, the theoretical models are under development and calculations with sufficient accuracy cannot be performed yet. For such atoms, experimental data are of major importance in practice. Presently, one of the most accurate methods to determine transition probabilities is the use of radiative lifetimes in combination with branching ratios.

Since the advent of lasers, they have been widely developed and explored in studies of atoms [19-20], including lifetime studies. Even high precision methods for lifetime measurements were demonstrated [21-23]. New applications of lasers were developed in different fields. In particular, the laser spectroscopy methods have been applied for studies of atoms in the UV/VUV (100-300 nm) spectral region (See, e.g. [24,25]). Nonlinear methods [26] have been used to transform tunable visible laser radiation into UV/VUV radiation. Experimental arrangements for time-resolved laser spectroscopy in the VUV spectral region have been developed at the Lund Laser Center [27]. There a large number of lifetime measurements have been performed using short-wavelength laser excitation [28].

2. Experimental methods

Pulsed laser excitation of evaporated substances and subsequent fluorescence decay detection have been used in the studies reviewed here. In our opinion this is the most convenient method for lifetime measurements in the UV/VUV spectral range. The principal scheme of an experimental set-up is shown in Fig.1. It has been slightly modified in various measurements, but the main parts are the same and correspond to the three tasks: atomization, excitation and detection. (The excitation laser pulse has to be of short duration and in the UV/VUV region.) Each of these tasks are discussed more in detail below.

2.1 Excitation

Until recently, the excitation of atoms in collisions with fast beams of electrons or in collisions of a fast beam of ions with a thin foil, was the only efficient way to populate highly excited states corresponding to UV/VUV transitions. Such methods have been employed in lifetime measurements of many atoms and highly charged ions [30-33]. But the non-selective excitation and possible influences of cascade decays are serious disadvantages of such methods. The excitation by laser light is selective. In quasi cw measurements using, e.g., the phase shift method, the measurement result becomes dependent on the laser power [34]. The pulsed laser excitation does not have such disadvantages.

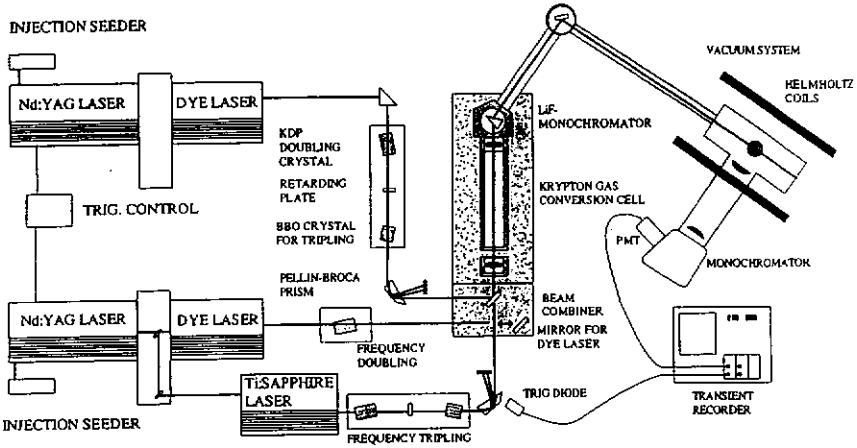


Figure 1. Experimental set-up for time-resolved laser spectroscopy (From Ref. [29]).

2.1.1 Short pulse generation

Normal pulsed lasers, chopped cw laser radiation, or mode-locked dye lasers can be used for the generation of the light pulses. The output powers of the cw and mode locked lasers are rather low and they can be applied only in the visible or, after frequency doubling, in the UV spectral region [35]. They can be used for lifetime measurements with high accuracy [36] Only radiation from pulsed lasers can be transformed down to the VUV spectral region by using non-linear effects.

Excimer or Nd:YAG-pumped dye lasers were used in many studies. The typical pulse duration is about 10 ns, sufficiently short for a large number of measurements (Fig. 2.).

In cases where shorter lifetimes were investigated, the experimental set-up has been modified. The simplest solution is to use a short-pulse nitrogen laser for pumping of the oscillator, while the amplifiers are pumped in the usual way; see Fig.3. The main problem of this scheme is the trigger jitter in the pump lasers, which leads to a non-perfect time overlap of oscillator and amplifier duty cycles, and therefore the outgoing radiation exhibits large fluctuations in intensity. Another way is to apply frequency mixing of the pump laser radiation with the dye laser radiation. The resulting output pulse from a nonlinear crystal is depending on the time overlap of the two input pulses, but there is no jitter. By delaying one pulse with respect to the second one, the pulse lengths can be shortened.

An alternative way is to use a ps laser pumping a distributed feedback dye laser. Such a system with two amplifier stages is shown in Fig. 4. By using a quenched distributed feedback system short pulses can be obtained even with a ns pump laser [39].

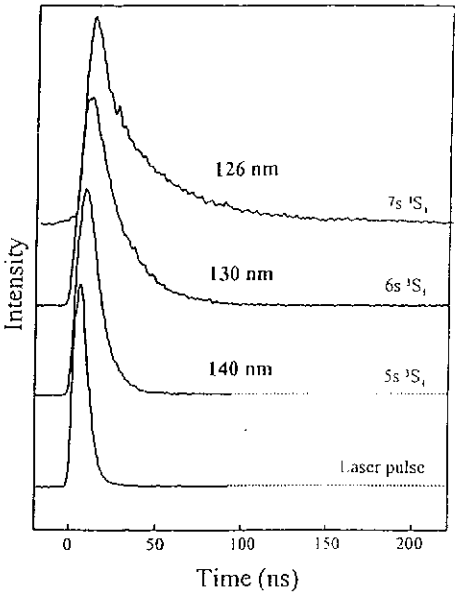


Figure 2. Recordings of fluorescence light transients for sulphur excited states and recording of scattered light from the laser pulse (From Ref. [37]).

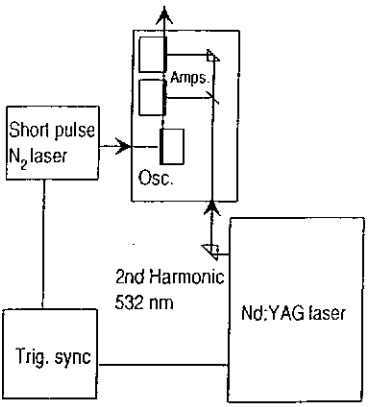


Figure 3. Experimental set-up for short-pulse generation using a nitrogen pump laser (From Ref. [38]).

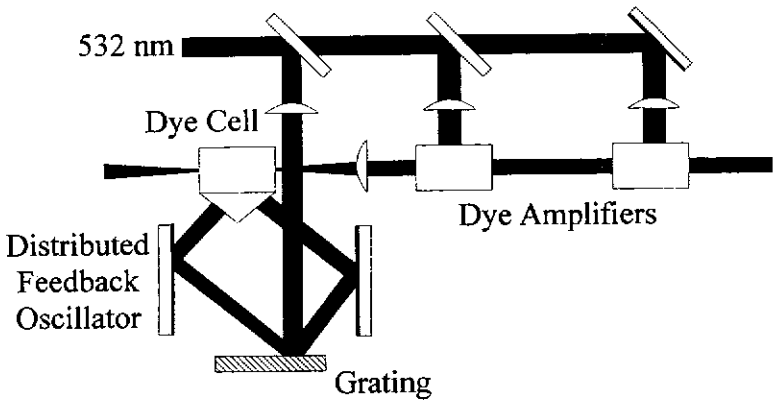


Figure 4. Experimental set-up for short- pulse generation by a distributed feedback dye laser (From Ref. [25]).

2.1.2. Generation of tunable UV/VUV laser radiation.

In early laser spectroscopy experiments the ions were shifted into resonance with lower energy photons through Doppler tuning, by changing the velocity of ions in a fast beam [23,40]. Presently, tunable laser radiation in the UV/VUV spectral region can be obtained from visible radiation by using the following nonlinear phenomena: harmonic generation and frequency mixing in crystals, stimulated anti-Stokes Raman scattering in gases and four-wave mixing in gases. Such generation down to the LiF absorption cut-off is illustrated in Fig. 5. Even shorter wavelengths can be reached by high-order harmonic generation in gases (See, e. g. [24,25]).

Nonlinear crystals are normally used for frequency doubling and mixing down to 200 nm, where the oxygen bands in air start to absorb. The β-barium borate crystal has a 189 nm cut-off, which is determined by the absorption in the crystal itself. Practically it is used down to about 200 nm, where the radiation is absorbed by oxygen in air. For shorter wavelengths the oxygen has to be removed from the path of the laser radiation.

Coherent radiation can be shifted towards shorter wavelengths applying stimulated anti-Stokes Raman scattering in gases. Particularly, the hydrogen molecule has the largest vibrational splitting corresponding to 0.5 eV. By using the second anti-Stokes component it is possible to shift 200 nm radiation down to 170 nm.

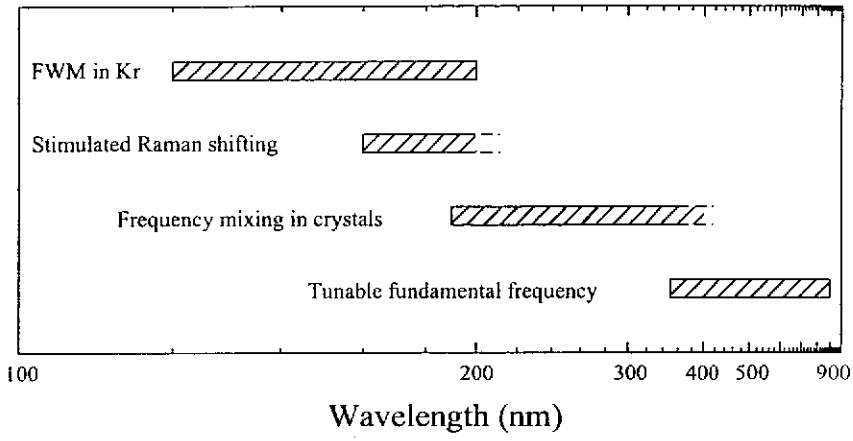


Figure 5. Different methods used for efficient generation of short-wavelength radiation down to the LiF cut-off (From Ref. [41]).

Shorter wavelengths can be reached through sum-difference four-wave mixing in gases. Two lasers are used, one of them in resonance with a two-photon transition in an atom, and the second one tunable. The resulting radiation has a photon energy defined by the difference between the two-photon energy and the tunable photon energy. In principle, using the resonant two-photon transition in krypton at 212.55 nm, any wavelengths longer than 106 nm could be obtained. The shortest obtainable wavelength is defined by the longest wavelength produced by the tunable laser, and it is about 120 nm if a Ti:sapphire or a dye laser is used.

An alternative to previously mentioned methods is high-order harmonic generation in gases. The harmonics are produced by focusing intense laser radiation into a jet of rare gases. Presently, the shortest wavelength obtained is in the order of 7 nm [42,43]. By using tunable dye or Ti:sapphire lasers tunable radiation at XUV and VUV wavelengths can be generated. An alternative way to obtain continuously tunable radiation is to mix high-power fixed frequency radiation with lower-power tunable radiation [44,45]. However, intense and short-pulse lasers have to be used for the generation of the really high harmonics. In spectroscopic applications the line-widths achieved with short-pulse lasers are frequently too large (Heisenberg uncertainty). Then laser systems with longer pulse duration are more useful for spectroscopy.

2.2 Detection

The fluorescence decay following pulsed laser excitation is normally recorded by a transient digitizer. This is a more efficient way to record decay curves in comparison with box-car integration or delayed-coincidence techniques. The fluorescence light is collected perpendicularly to the plane of the laser and atomic beams by quartz or lithium fluoride lenses and is selected by a monochromator or a filter before reaching a photomultiplier tube. Transients are stored in the memory of a fast digital oscilloscope. The time resolution is defined by the rise time (3 ns) of the photomultiplier tube. For faster measurements, a micro-channel plate has been applied. The accuracy of this detection scheme is limited by the linearity of the detectors, which have to measure both high and low intensity signals separated in time by short intervals.

When a very high sensitivity is needed the pump/probe technique using photo-ionization was employed [46]. The atoms were excited by the pump beam and then a delayed probe beam was applied to ionize them from the excited state. The ion signal versus delay time is detected. Advantages of this technique are: a high sensitivity and a time resolution limited only by the duration of the pump and probe pulses, but not by the time resolution of the detector.

2.3. Atomization

Different methods have been used for producing free atoms. For lifetime measurements atoms have to be studied in conditions, where the interaction with the environment is very low. Usually, that is inside a vacuum system. The easiest way of producing free atoms in the vacuum system is by leaking in a gas consisting of the atoms needed [47,48]. But only a few elements are available in the gas phase at room temperature, therefore, the thermal evaporation from an oven inside the vacuum system is often used. Many materials evaporate as molecules and then the dissociation of molecules is a special task. In some cases, the laser light used for excitation serves for dissociation, too [47-49]. When a small amount of substance is available or if it is toxic, sealed-off cells were used in the measurements [50,51]. However, they have several disadvantages: the presence of rest gas in the cells, a large amount of stray light from laser light scattering in the cell windows and the wavelength range of the applied radiation is limited by absorption in the cell windows. When they are made from quartz the limit is around 160 nm.

If the material is difficult to vaporize by thermal heating, bombardment with electrons and extraction from the discharge can be applied in an effusive hollow-cathode arrangement [52,53]. An alternative way is to use focused laser light to produce a plasma containing atoms and ions of the species under investigation. A conventional target consists of a disc, with a surface of the material to be studied [54-56]. The disc is rotated with the purpose to bring a new flat surface in the focus after each laser shot. For some materials the fabrication of such a disk is a rather complicated task and then a powder in a container can be used (Fig. 6.) [57-59]. The container is shaken at a high frequency and the surface of the powder is brought back in the light focus before the next laser pulse. A few μs after the laser hits the target no emission from the plasma itself is observed, and the excitation laser pulse can be applied (Fig. 7.).

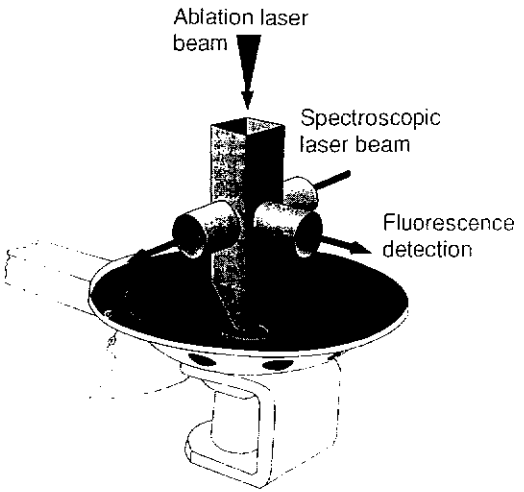


Figure 6. Arrangement for laser-produced plasma generation from a powder (From Ref. [57]).

A sufficient number of atoms has to be produced to be able to detect a fluorescence signal. On the other hand, when the density of atoms is too large, the collisional quenching interferes with the radiative lifetime. Normally, a Stern-Vollmer plot is drawn and the lifetime extrapolated to zero concentration is taken as the radiative lifetime (Fig. 8.). Extrapolation is simple when using a cell containing the atoms.

When a thermal beam or a laser-produced plasma is applied, it is much more difficult to evaluate concentrations. Then the lifetimes are measured at different experimental conditions in order to observe possible influences of collisions on the measured lifetimes; see Fig. 9

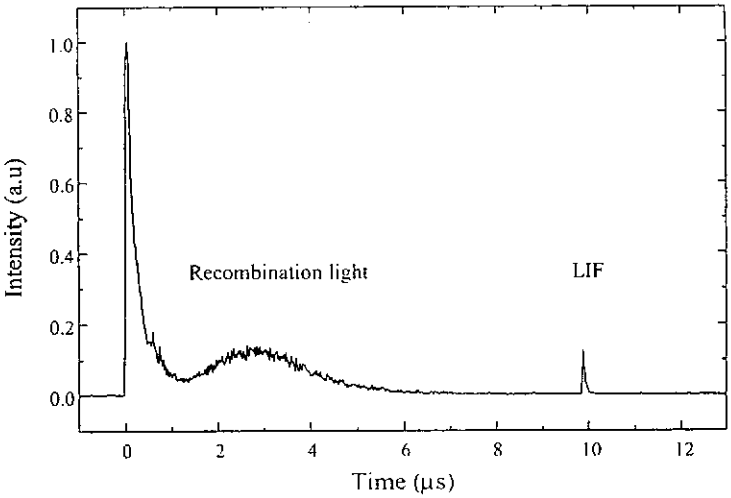


Figure 7. Detected light intensity at 126 nm from a sulphur plasma as a function of the time after the ablation pulse. The signal at about 10 μs is laser-induced fluorescence from the excited state. The signal before 6 μs is due to recombination light from atoms and ions. (From Ref. [37]).

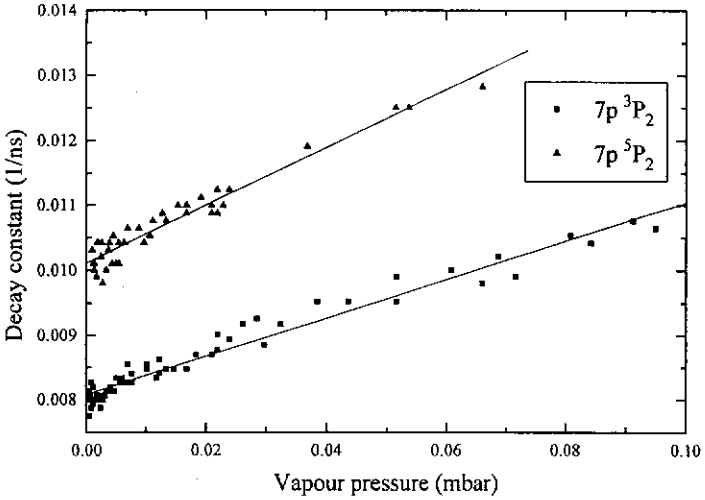
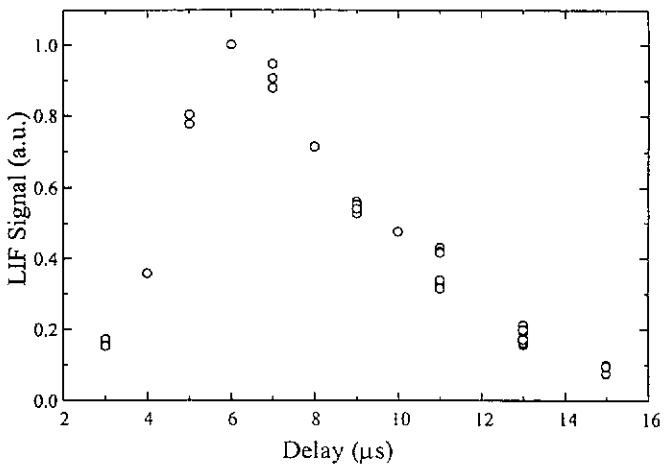


Figure 8. Stern-Vollmer plot for tellurium 7p $^5P_2$,$^3P_2$ state lifetime determination (From Ref. [49]).

a)



b)

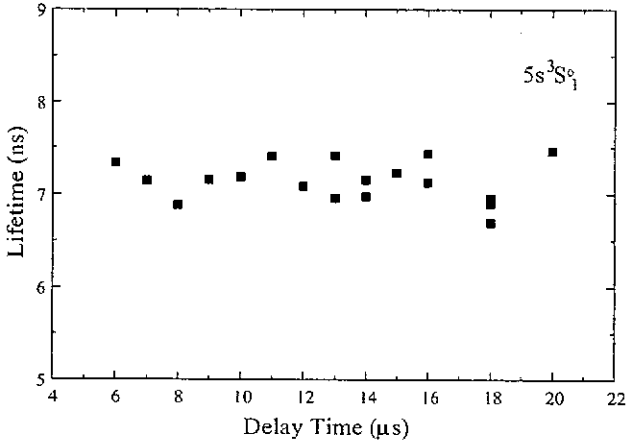


Figure 9. The time dependence of the fluorescence light intensity and decay time for a laser-produced sulphur plasma: a) Laser-induced fluorescence intensity from the 7s $^3S^0_1$ state as a function of the delay time for the VUV laser pulse with respect to the ablation pulse. b) Measured radiative lifetime of the 5s $^3S^0_1$ state as a function of the delay. (From Ref. [37]).

2.4. Sources of possible errors

We already mentioned two possible sources of errors: a non-linearity of the detector, and the collisional quenching of excited states. A non-linearity of the detector is intensity dependent; by recording fluorescence signals at different intensities the influence of possible saturation of the detector can be observed. Radiation trapping is one of the processes changing the number of atoms in excited states. It is a density dependent process and may be taken into account by carrying out measurements at different densities. Another problem is possible blends from other transition lines. This has to be checked very carefully if interference filters are used and molecular lines are overlapping with detection lines. The best way to solve this problem is to use different decay channels for the detection. Influences from quantum beats have been reduced by applying an additional magnetic field over the interaction region. For the higher Rydberg states a possible influence of blackbody radiation has to be accounted for [60].

3. Results

High lying excited states are accessible using UV/VUV radiation. Various types of levels can be excited: Rydberg states for large groups of atoms, resonance states for another group of atoms and ions in the first ionization stage. For many of the states the first lifetimes where measured by the LTH group. These results have served as test data for theoretical calculations. In some cases a comparison between experimental results obtained with different methods was possible and thus serving as a test of the methods. For different atoms and ions the motivation for lifetime measurements were slightly different.

In a large group of papers lifetime measurements of irregularities in long Rydberg sequences were performed for elements Group 2a (Mg I, Sr I) [61,62], Group 3a (Al I, Ga I, In I) [63-65], Group 1b (Cu I, Ag I, Au I,) [66-71] and Yb I [72], Zn I [73] and Bi I[74]. For the Group 2a and 3a elements non-perturbed S sequences and perturbed D sequences were observed. The P sequence of group 1b atoms was found to be strongly perturbed, while S and D sequences were following a cubic rule. Lifetimes of P sequences in ytterbium also show strong influence of a perturbation, which was confirmed by g_j factor measurements. This initiated further Multi-channel Quantum Defect Theory analysis. In zinc, a deviation was observed between measured and calculated data. In bismuth rather good agreement with a cubic rule was observed, with the exception for one state, for which the hyperfine structure was also found to be perturbed.

Measurements of lifetimes in Gd II, Ru II, Pt II, Pd II, As I, P I and S I [53,57,75,76,50,59,58,37] were motivated by a strong interest from

astrophysicists. The aim was transition probability values. The measurements were performed on astrophysically interesting states, and data obtained were used for the evaluation of the abundance of elements in astrophysical objects. Experimental and theoretical branching ratio data were used for evaluation of transition probabilities. The possible astrophysical applications as well as the existence of experimental branching ratio data, suggested experiments on selenium and tellurium. For bismuth a revision of transition probability data was performed for more than 20 lines [74].

In some experiments new experimental schemes were applied. In the lifetime measurements of O I, a two-photon excitation in a first step also inducing molecular dissociation, followed by a second excitation step was demonstrated [47]. That allowed to excite atoms to levels otherwise corresponding to direct groundstate transitions with wavelengths <100 nm. In later experiments on N I even higher levels were reached [48]. In an experiment on C I two-photon excitation and lifetime measurements were achieved with detection through cascades via a short-lived state [55], a technique also successfully later applied on S I and P I [58,59]. The application of four-wave mixing was demonstrated in experiments on Mg I [29] and later used for studies on Au I, Zn I and S I [71,73,37]. The application of a new picosecond laser system based on a mode-locked Nd:YAG and a distributed feedback dye laser was demonstrated in paper [71].

4. Future perspectives.

Further progress in experimental techniques will allow lifetime measurements in neutral and singly ionized atoms to be extended to even shorter wavelengths and lifetimes. Any free neutral and singly ionized atom can be produced in a laser-generated plasma. For short-lived states, the pump-probe technique allows to overcome the problems with detector response time. A distributed feedback laser pumped by picosecond pulses from a mode-locked Nd:YAG laser can serve as a light source.

In astrophysics transition probabilities are needed and branching ratios have to be used in conjunction with lifetime data. In the VUV region accurate branching ratio measurements are complicated due to difficulties in calibration of the spectral sensitivity [77]. One way to overcome this problem is to perform transition probability measurements in absorption for lines with a common lower level. This approach, called the Ladenburg method, was already demonstrated in the UV part of the spectrum [78,79]. In view of the strong ongoing effort in space-based short wavelength astronomy there is a strong motivation for future studies of transition probabilities in the UV/VUV part of the spectrum.

Acknowledgments

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This paper is dedicated to Professor Ingvar Lindgren on the occasion of his 65th birthday.

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